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Electromagnetically induced transparency with localized impurity electron spins in a semiconductor

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Chapter 4

Electromagnetically induced transparency as probe for nuclear spin polarization around donor-bound electrons in GaAs

Abstract

We study how electromagnetically induced transparency (EIT) with donor-bound electrons in high-purity n -GaAs can be applied as a probe for the dynamics of nuclear spin polarization. The localized donor electrons have hyperfine interaction with surrounding nuclear spins, and probing the electron spin splitting with EIT thus reflects the nuclear spin polarization. We also apply that scanning spectroscopy around the EIT resonance predominantly acts as optical pumping on the electron spin, which provides a bidirectional pump for dynamic nuclear polarization (DNP). EIT spectroscopy can thus follow DNP both during build-up and its relaxation, and we characterize how such EIT probing can be tuned between a drive and weakly invasive probe for nuclear spin polarization.

This chapter is based on Ref. 4 on p. 127.

4.1 Introduction

Localized electrons in semiconductors have received attention in recent years for the possibility of preparation, manipulation and detection of the electron-spin state. Examples of these systems are quantum dots [1, 2, 3, 4], rare earth doped crystals [5, 6], color centers [7] and donors in semiconductors [8, 9, 10]. A common challenge in these systems is to control their interaction with the crystal in which they are embedded. Of particular interest is counteracting the coupling to thermally fluctuating variables of the environment, which generally is detrimental to the electron-spin coherence. In the group III-V semiconductors, the nuclei possess non-zero spin, whose fluctuating polarization interacts with the localized electron spin via hyperfine interaction [11, 12, 13, 14, 15]. This results in the lowered value of the spin dephasing time, T_2^* . With emerging research showing the possibility to enhance T_2^* by controlling the nuclei, it is important to be able to measure this nuclear-spin environment and exert control over it.

Here we present measurements on the nuclear spin environment of donor-bound electrons (D^0) in gallium arsenide (GaAs) using the all-optical technique of electromagnetically induced transparency (EIT). We show how EIT can be used to precisely measure the effect of a polarized nuclear spin environment on the donor-bound electrons. Further, we explore how the EIT-control lasers themselves can play a role for controlling the nuclear spin environment via dynamic nuclear polarization (DNP), which is relevant for making EIT an effect that can improve itself by suppression of the detrimental influence of disordered nuclear spin orientations [16]. Quantitative measurements of the invasive behavior of EIT driving and interplay with DNP are presented. An advantage of this method, over nuclear magnetic resonance, is that it probes nuclear spin polarization locally at the position of the D^0 wave function.

4.2 Material

We use n -doped GaAs with a silicon doping concentration of $\sim 3 \times 10^{13} \text{ cm}^{-3}$. At 4.2K, the D^0 electrons are localized around the silicon atoms. The D^0 electron wave functions have a hydrogenic energy spectrum, thus forming a solid-state analog of an alkali atom inside the crystal. With an effective Bohr radius of 10 Å they are well separated from each other at the mentioned concentration, forming a homogeneous ensemble localized electrons. These systems have an optically excited state, the neutral donor-bound exciton (D^0X), consisting of two electrons

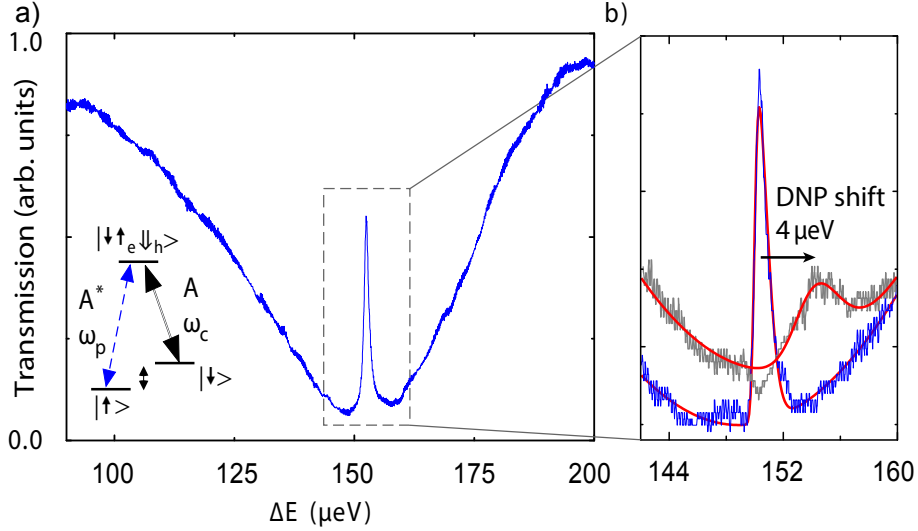


Figure 4.1: (a) Electromagnetically induced transparency (EIT), observed as a narrow peak in the transmission spectrum from a two-laser experiment on an ensemble of the donor-bound electrons in n -GaAs. The ensemble is addressed in a Λ -configuration (see inset), using the two Zeeman-split levels of the donor electrons and the lowest optically-active level of the donor-bound exciton complex. The control laser is fixed at the A -transition while a probe laser scans across the A^* -transition, the photon-energy difference, ΔE , between the lasers is along the horizontal axis. Intensities are 3.7 W cm^{-2} and 0.18 W cm^{-2} for control and probe respectively. (b) Magnified view of the central part of the spectrum in (a), showing the EIT peak before (blue) and after (gray) inducing nuclear spin polarization. The EIT peak in the gray trace (arrow) shows a shift and is broadened. Red lines are Gaussian fits to the data (see main text for details) yielding peak positions of $(150.47 \pm 0.01) \mu\text{eV}$ and $(154.47 \pm 0.05) \mu\text{eV}$ respectively

and a hole bound to the donor. To perform EIT experiments it is required to have a three-level system. This is achieved by applying a 6.5 T magnetic field from a superconducting coil to lift the spin degeneracy in the D^0 and the D^0X . The Zeeman split D^0 -spin states and lowest energy D^0X -state (of levels that can be easily observed) form a three level Λ -system as shown in the inset of the Fig. 4.1(a), where the D^0X -state is denoted as $|\uparrow\downarrow_e\downarrow_h\rangle$. Following Refs. [17, 9] we denote the optical transition from $|\downarrow\rangle$ by A and from $|\uparrow\rangle$ by A^* .

4.3 Spectroscopy method

For our experiments we use the setup as described in Ref. [18] where two laser beams co-propagate through the sample and are collected immediately behind the sample on a photodiode. The spot diameter of both lasers at the sample is approximately $10\ \mu\text{m}$. One of the laser beams is modulated by a chopper at 6 kHz and we isolate the modulated part of the total transmission signal by lock-in detection (for details see [19]). Measurements are performed in Voigt geometry (laser propagation perpendicular to magnetic field direction) such that the A - and A^* -transitions couple to V- and H-polarized light respectively. To perform EIT measurements one laser is kept resonant (denoted as the control laser) and the frequency dependent transmission is recorded while scanning the other laser (denoted as probe laser) over the other transition. The $55\ \mu\text{eV}$ broad (FWHM) absorption dip is observed in Fig. 4.1(a) with a sharp, $1\ \mu\text{eV}$ wide, enhanced transmission window inside it due to destructive quantum interference. This EIT resonance appears exactly at the two-photon Raman resonance condition where the energy difference between the laser photons equals the electron spin splitting [20, 21].

4.4 Dynamic nuclear spin polarization by optical excitation

The nuclear spins at the D^0 centers can be polarized in a process known as dynamic nuclear polarization (DNP). This is an indirect method where first the electron spin is brought out of thermal equilibrium, which can be done by optical pumping techniques. Subsequently, angular momentum gets transferred to the nuclear spins in the environment through hyperfine interaction [15], creating a net spin polarization of the nuclei. The hyperfine coupling to (partially) spin polarized nuclei induces an effective magnetic (Overhauser) field felt by the electron spin, causing a change in the electron spin splitting. Here we use resonant optical excitation of either one of the Λ -system transitions to drive the DNP process. After such optical pumping the resulting Overhauser field causes a shift in the EIT peak position. This shift is shown in Fig. 4.1(b). A change in line shape is also apparent, this originates in the fact that we observe an ensemble of D^0 systems with a slight spread in spin splitting. The width of the EIT peak relates to this spread and results in the T_2^* of approximately 3 ns for the equilibrium case (blue line) [9, 10]. In the next section we comment on how this inhomogeneity is

reflected in the transmission signal.

4.5 Relation between EIT line shape and Overhauser field

The line shape of the EIT resonance contains information on the spin polarization of the nuclei near the donors. In two-laser spectroscopy the measured transmission of each of the lasers through the GaAs sample is determined by the amplitude transfer function $T(\omega_i, \Omega_i | \omega_j, \Omega_j) = \exp(i\omega_i n d / c \chi(\omega_i, \Omega_i | \omega_j, \Omega_j) / 2)$, where d is the thickness of the medium, n is the refractive index of GaAs and c is the speed of light, $i, j \in 1, 2$ with $i \neq j$ labels the laser frequencies ω and intensities expressed as Rabi frequency Ω , as also introduced in Fig. 4.2. The notation of separating the variables in T and χ by a vertical line is meant to indicate that we consider the transfer function with variables ω_i, Ω_i conditional on ω_j, Ω_j which then assume the role of parameters. The susceptibility χ is obtained by considering the polarization density of a medium filled with donors with concentration ρ , each represented as a Λ -system as described in Fig. 4.2. The dependence of the transmittance on the decay and dephasing parameters of the Λ -system is implicit.

The susceptibility is made up from the polarizabilities of individual systems β which can, and generally will, show small differences, thus forming an inhomogeneous ensemble. We focus on the inhomogeneity arising from the nuclear spin polarization. This inhomogeneity gives rise to a distribution of Overhauser shifts $P(\delta)$. Here the value of $\delta = p\delta_{\max}$ is proportional to the nuclear spin polarization $p \in [-1, 1]$, where δ_{\max} is the maximum shift set by the hyperfine interaction strength. For the donor electron in GaAs $\delta_{\max} = 24.5$ GHz (obtained from the maximum Overhauser field [22] via $\delta = g\mu_B B_n / 2\hbar$ with g-factor $g = -0.41$ [10]). We express the susceptibility for the medium accordingly as

$$\chi(\omega_i, \Omega_i | \omega_j, \Omega_j) = \rho \int P(\delta) \beta(\omega_i, \Omega_i | \omega_j, \Omega_j, \delta) d\delta. \quad (4.1)$$

The polarizability β exhibits an EIT resonance (β decreases as the transmission increases). When this resonance is narrow as compared to $P(\delta)$ (such that it can be approximated by a Dirac delta function), Eq. 4.1 implies that the transmission near EIT resonance has the shape of $P(\delta)$. The approximation is valid for the ensemble of D^0 electrons, because their measured individual decoherence time (at least (7 ± 3) μs [23]) implies a very narrow (sub MHz) EIT feature. This

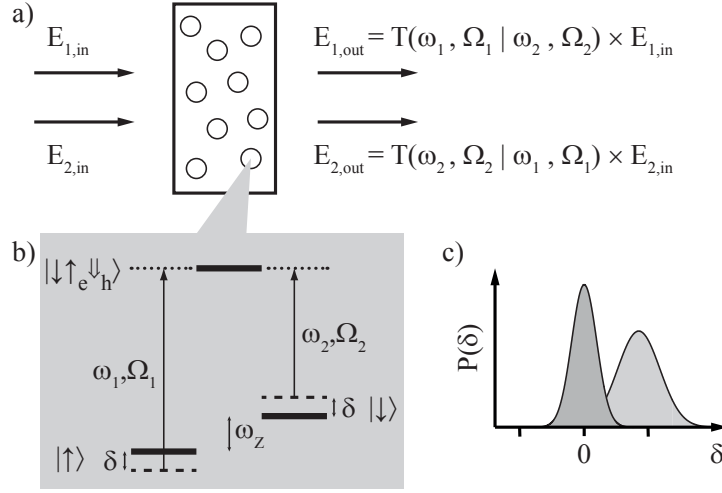


Figure 4.2: Transmission through a medium containing inhomogeneous Λ -systems. (a) The transmission of each of the two laser fields is determined by an amplitude transfer function, T , which is conditional on the intensity and frequency of the other laser when the lasers are both close to resonance with the transitions of a Λ -system. T follows from the susceptibility, which is an effective description of the medium and is in turn derived from the polarizabilities of the individual Λ -systems. One of these is depicted in (b). Nonzero nuclear spin polarization causes a shifting of the electron-spin states by $\pm\delta$. This changes the two-photon resonance condition. (c) The inhomogeneity in nuclear spin polarization is introduced through having δ governed by a probability distribution $P(\delta)$. Dynamic nuclear polarization can change the position and shape of the initial $P(\delta)$ (dark gray), such that the mean shifts away from zero (e.g. light gray area).

property is used to extract the average Overhauser field by fitting the data as shown in Fig. 4.1(b). A second order polynomial is used to fit the background (areas left and right of the peak) and a skew-Gaussian fit is used for the EIT resonance. The fitted curves are shown as red lines in Fig. 4.1(b), with peak positions at $(150.47 \pm 0.01) \mu\text{eV}$ for the case where the nuclear spins are at thermal equilibrium (blue data) and $(154.47 \pm 0.05) \mu\text{eV}$ for the case after DNP pumping (grey data). The error bounds indicate the 95% confidence interval of the least-squares Gaussian fit. In following figures we take the peak positions of such fits to extract the average Overhauser field, *i.e.* $(4.00 \pm 0.05) \mu\text{eV}$ for the case shown in Fig. 4.1(b).

From the fits to the blue and gray traces it is apparent that this EIT-based measurement is very accurate for extraction of the average spin splitting of the

D^0 electrons. The error of approximately one part in ten thousand is comparable to spin-noise measurements on similar samples [24]. For the case after optical pumping a broadening of the resonance is visible, resulting in a larger error bound for the fit. We attribute this to the fact that the pump induced DNP is intensity dependent, and the intensity of the laser field throughout the sample is not uniform because of the (radial) Gaussian spot and (longitudinal) Fabry-Pérot interference (see also our discussion on this in [16, 25]). Nevertheless it is possible to determine the average Overhauser field with high accuracy since the measured EIT spectrums directly reflect $P(\delta)$ (see above). In the following sections we use this method to investigate the build-up and decay of DNP in time.

4.6 Build-up of nuclear spin polarization

By taking EIT measurements during optical pumping of the electron spin we measure the build-up time of DNP. In this case the control laser, which is at higher intensity, causes the optical pumping leading to DNP (and we refer to it as the pump laser in this context). The experimental protocol to induce and simultaneously detect the DNP is shown in the inset of Fig. 4.3(b): After having the sample in the dark for tens of minutes (fully relaxing the DNP effects) the pump laser is switched on at $t = 0$, at an intensity of 6 W cm^{-2} . While the weak probe laser is repeatedly scanned to take EIT spectra, the pump laser fulfills the role of the control laser required to make the EIT resonance visible. The pump is optically exciting the A^* -transition and spectra are recorded with a probe laser of intensity 0.1 W cm^{-2} by scanning across the A -transition. The transmission spectra, Fig. 4.3(a), show how the position of the EIT resonance changes as a function of time. The spectra are plotted on top of each other with an offset along the vertical axis for clarity. The time difference between two consecutive spectra is 65 s, which is the duration of a single scan. The shift in the peak position indicates a change in the electron spin splitting. Since this is observed at a constant external magnetic field it means the control laser acts as a pump to induce DNP in n -GaAs at the donor sites. The peak positions are extracted from Fig. 4.3(a) and plotted as a function of time in Fig. 4.3(b). This shows the build-up of the Overhauser field with a time constant $\tau_{\text{build}} \approx 340 \text{ s}$, obtained by a least squares fit to the exponential association function $\Delta E = \Delta E_{\text{max}}(1 - \exp(-t/\tau_{\text{build}}))$. This way of monitoring the Overhauser field during its build-up can be used to study the evolution of the nuclear field and its time scale.

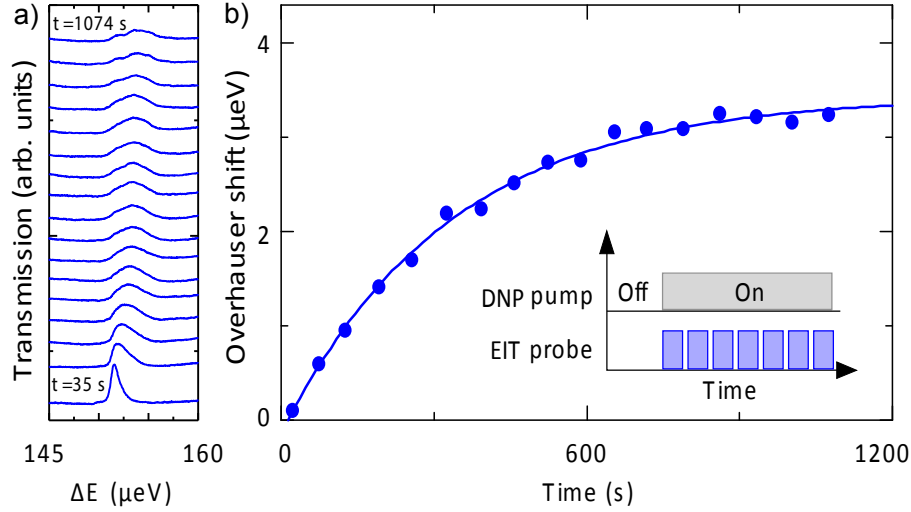


Figure 4.3: (a) The effect of optical pumping on the EIT peak shape and position (traces offset for clarity), in an experiment where a strong laser (6 W cm^{-2}) drives the A^* -transition, acting simultaneously as pump laser for DNP and control laser for EIT probing. A weak probe laser (0.1 W cm^{-2}) periodically scans the A -transition (see time labels). Time $t = 0$ is defined as the moment where the pump laser is switched on after the sample has been in the dark (see inset of (b)). (b) The Overhauser shift as derived from the data in (a) as a function of time (dots). The solid line is a fit for exponential build-up, with a time constant $\tau_{\text{build}} = 340 \text{ s}$.

4.7 Decay of nuclear spin polarization and bidirectionality of DNP

Detection of nuclear polarization by EIT can also be used to monitor decay of nuclear spin polarization after it first has been induced by optical pumping. The experimental protocol is explained in the Fig. 4.4(c) inset where, in a first step, the pump laser excites one transition of the Λ -system at intensity 6 W cm^{-2} for 30 minutes to induce DNP. Then, in a second phase, the probe and control laser are used to perform EIT measurements, with the control laser addressing the A -transition. This experiment is performed in two distinct ways, one by optical pumping on the A^* -transition (Fig. 4.4(a)) and another by optical pumping on the A -transition (Fig. 4.4(b)), as indicated by the displayed energy level schematics.

After optical pumping on the A^* -transition, subsequent EIT measurements are collected with the control and probe laser intensity of 2.37 W cm^{-2} and 0.1 W cm^{-2} respectively. The first spectrum in the Fig. 4.4(a) shows the EIT

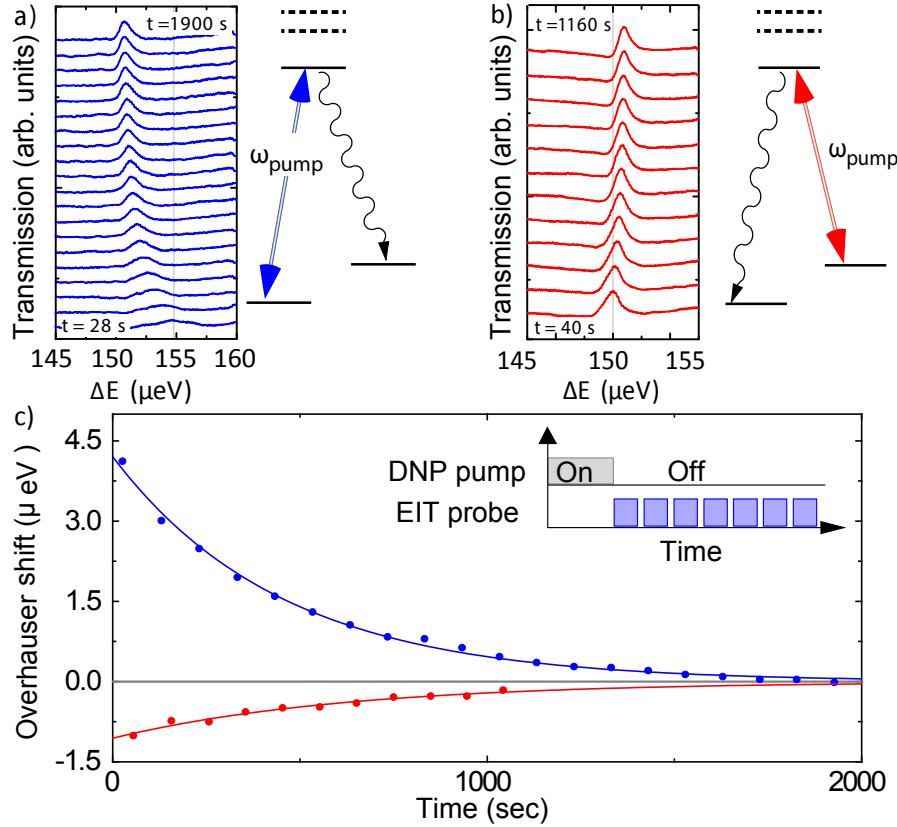


Figure 4.4: Bi-directionality and decay times of the induced Overhauser shift. (a) Evolution of the EIT peak shape and position after a single laser (6 W cm^{-2}) has been driving the A^* -transition for 30 min for inducing DNP (traces offset for clarity). Time $t = 0$ is defined as the moment where the DNP driving is stopped, and EIT probing starts with the control (2.37 W cm^{-2}) on the A -transition and scanning probe (0.109 W cm^{-2}) on the A^* -transition (see inset of (c)). (b) Similar to (a), EIT data taken after pumping the A -transition (6 W cm^{-2}) for 30 min. The EIT probing is realized in the similar manner as for (a) by keeping control to probe ratio the same. (c) The Overhauser shift as derived from the data in (a) and (b) as a function of time (dots). The solid lines are fits for exponential decay, with time constants τ_d of 600 s (after pumping A -transition) and 480 s (after pumping A^* -transition).

peak shifted to higher energy as compared to its equilibrium position in the last (upper) trace. The time difference between two consecutive spectra is 93 s, which is the time required to collect one scan. The sequence shows the evolution of the EIT peak position and shape towards equilibrium. It shows that the nuclei become polarized in the direction of the applied magnetic field when the A^* -transition is pumped, because spin splitting is enhanced. This implies the optical pumping creates an Overhauser field along the applied magnetic field.

For the measurement in Fig. 4.4(b), the pump laser was resonant with the A -transition. The same EIT measurement protocol is followed as for Fig. 4.4(a). The control and the probe intensity are 4.7 W cm^{-2} and 0.2 W cm^{-2} respectively during the EIT measurements. These spectra are plotted in the same manner as in Fig. 4.4(a). The time difference between recording two consecutive spectra is 93 s. In this sequence, the first EIT peak at time $t = 40 \text{ s}$ appeared at lower energy as compared to the Zeeman energy at the applied magnetic field. It shows that the Overhauser field opposes the applied magnetic field when the A -transition is pumped.

In Fig. 4.4(c) the peak positions from Fig. 4.4(a) and Fig. 4.4(b) are plotted as a function of time exhibiting exponential decay curves that can be fitted with time constants of 600 s and 480 s. The intensity of the pump laser was equal for acquisition of both data sets. It shows that the DNP can be induced in either parallel or antiparallel to the external magnetic field, depending on the transition the pump addresses. This is consistent with the effect of DNP originating from an induced out-of-equilibrium spin polarization of the electron. The magnitude of the induced Overhauser shifts and their decay times are not symmetric. For the induced Overhauser field we attribute this to the fact that the electron spin population at thermal equilibrium is $n_{\uparrow}/n_{\downarrow} = \exp(g\mu_B B/kT)$ (Boltzmann factor), which at 4.2 K and 6.4 T implies that approximately 60 % of the population resides in the ground state. Hence, a greater difference (with respect to thermal equilibrium) in spin population can be created by pumping the A^* -transition than by pumping the A -transition. The observed relaxation of the nuclear spin polarization at the D^0 electrons can be ascribed to diffusion of nuclear spin polarization away from the donor site [22], that it is different for the different directions of Overhauser field is a peculiar observation. However, it should be taken into account that when taking EIT scan with the control laser on the A -transition, the electron spin is polarized in the same way as for the optical pumping performed before acquiring the data in Fig. 4.4 b. Hence, the EIT scans themselves induce DNP, making the decay appear slower. This invasiveness of EIT is analyzed in

the next section.

4.8 Invasive character of EIT as a probe

Optical excitation of the transitions of the lambda systems cause nuclear spin polarization. This implies that it is possible to disturb the nuclear spin environment during EIT measurements. To investigate the invasive behavior of EIT measurements the control laser (with intensity 2.16 W cm^{-2}) is kept in resonance with A -transition and a transmission measurement is performed using the probe with intensity 0.216 W cm^{-2} across the A^* -transition. The recorded transmission spectrum is shown as the red line in Fig. 4.5(a). When the control is instead resonant with the A^* -transition (at 1.65 W cm^{-2}) a transmission measurement is recorded using the probe with 0.093 W cm^{-2} scanning across the A -transition (blue line in Fig. 4.5(a)). The EIT resonances are located at $150.87 \mu\text{eV}$ (red) and $151.66 \mu\text{eV}$. The difference of $0.79 \mu\text{eV}$ in the EIT peak positions implies that the electron spin splitting is influenced by the sign of the electron spin polarization during EIT scans.

Since the EIT probing causes unwanted nuclear spin polarization, we quantify its effects by measuring the decay times of the nuclear spin polarization in the dark as well as illuminated. DNP is induced in the same way as for Fig. 4a. The EIT peak position is plotted as a function of time, shown in Fig. 4.5(b) (black circles). This result is compared with the situation where the system is kept in the dark. To achieve this, optical pumping is performed for 30 minutes, followed first by EIT measurement immediately after. After this initial EIT measurement the system is kept in the dark by blocking all the lasers. Then another EIT measurement is performed after an elapsed time (indicated on the horizontal axis Fig. 4.5(b)). The EIT peak positions are plotted as a function of time as the grey data triangles in Fig. 4.5(b). For this data set the decay takes place almost entirely in the dark and we expect the invasiveness of the EIT measurement to be minimized. Exponential fit to the black data points gives decay time constant of 600 s and exponential fit to the grey data points gives decay time constant of 260 s, the latter is indeed faster as expected.

To be able to monitor the decay of nuclear spin polarization by more closely, it is required to eliminate the effect that an out-of-equilibrium spin polarization during the EIT scan induces DNP itself. To counter this, ideally, the intensities of control and probe laser should be tuned such that the spin polarization during the scan equals the equilibrium spin polarization. We show how this situation can

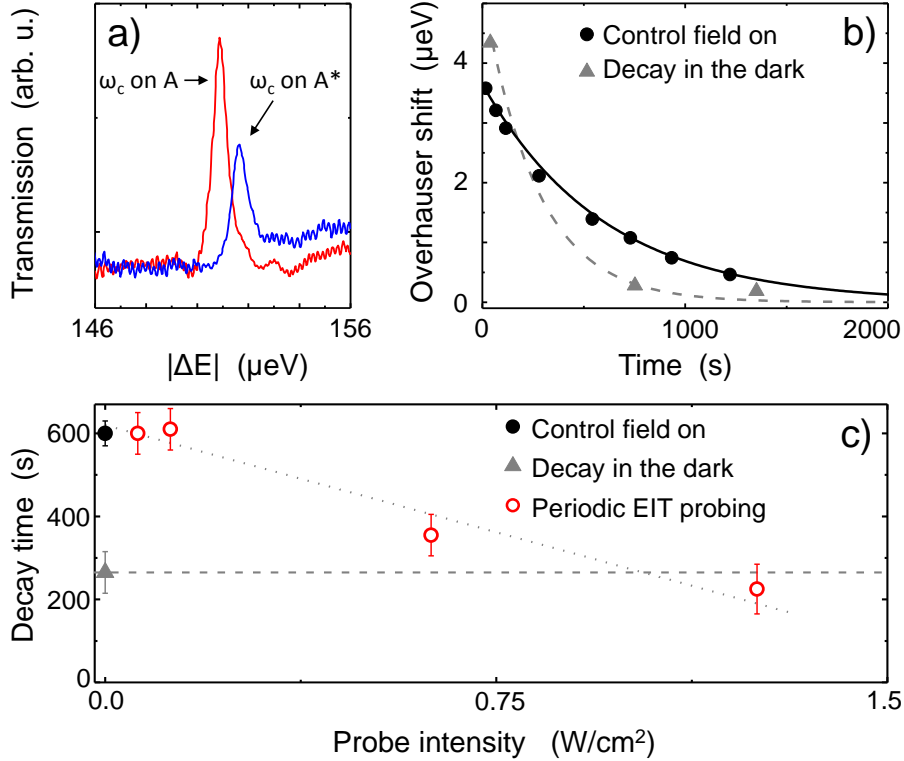


Figure 4.5: (a) Studies of the invasive character of EIT as a probe. (a) The difference in EIT peak position for the control (photon energy $\hbar\omega_c$) on the A - and A^* -transition, taken with similar intensities for the control (2 W cm^{-2}) in the two cases, and weaker probe lasers (see main text). (b) Decaying Overhauser shifts, derived from EIT scans taken after pumping the A^* -transition (6 W cm^{-2}) (dots). The decay with the sample in the dark between initial and subsequent EIT probing goes faster than decay with an EIT control laser continuously on the A -transition (EIT probing in both cases with control on A). Lines are fits for exponential decay, yielding time constants of 280 s (in dark) and 600 s (control on). (c) Time constants from exponential-decay fits for the data of (b), and similar data from periodic EIT probing with a control of 6 W cm^{-2} on A - and a probe of varying intensity (see horizontal axis) scanning over the A^* -transition (dashed/dotted lines are a guide to the eye).

be approached empirically by varying the intensities of the optical fields for the EIT measurements. The measurement protocol of Fig. 4.5(b) is repeated. And, while all other factors remain constant, the intensity of the probe laser is varied from 0.06 W cm^{-2} to 1.5 W cm^{-2} . The resulting decay times are represented by red data points (black and grey data points correspond to panel (a)). It is observed that the relaxation time as the ratio of the probe to control intensity is approximately one, is identical to the relaxation time when system is kept in the dark in between the measurements. In this regime EIT can be treated as a weakly invasive tool to probe the nuclear dynamics. An alternative approach to this problem where both control and probe intensities are lowered to be incapable of inducing DNP is not possible. This is because to observe EIT it is required that the control laser coupling has a Rabi frequency exceeding the excited state decay time.

4.9 Conclusion

To be able to create internal magnetic Overhauser fields in a material is useful for spintronics applications. Monitoring the dynamics of nuclear spin polarization associated with this gives valuable insight in the electron-nuclear spin dynamics. The all-optical method presented here can be used as a weakly invasive method to locally probe these fields. It compares in accuracy with other all optical techniques, such as the recently demonstrated measurement of Overhauser fields by spin noise measurement [26]. An important advantage is that the technique used in our work, using two CW lasers in EIT configuration, already fulfills the requirements for techniques of light storage on spin states [27, 28, 29] and coherent control of electron spin states [30] which are important precursors for achieving quantum information applications with spins and photons.

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